

220355US0PCT

U.S. APPLICATION NO. (IF KNOWN, SEE 37 CFR

10/089065

INTERNATIONAL APPLICATION NO.

PCT/EP00/09630

INTERNATIONAL FILING DATE

2 October 2000

PRIORITY DATE CLAIMED

5 October 1999

TITLE OF INVENTION

CHEMICAL AND/OR PHYSICAL TREATMENT OF MIXTURES WHICH CONTAIN AT LEAST ONE
CHEMICAL COMPOUND HAVING AT LEAST ONE ETHYLENICALLY UNSATURATED GROUP

APPLICANT(S) FOR DO/EO/US

MUELLER-ENGEL Klaus Joachim et al.

Applicant herewith submits to the United States Designated/Elected Office (DO/EO/US) the following items and other information:

1. ☒ This is a **FIRST** submission of items concerning a filing under 35 U.S.C. 371.
2. ☐ This is a **SECOND** or **SUBSEQUENT** submission of items concerning a filing under 35 U.S.C. 371.
3. ☒ This is an express request to begin national examination procedures (35 U.S.C. 371(f)). The submission must include items (5), (6), (9) and (24) indicated below.
4. ☒ The US has been elected by the expiration of 19 months from the priority date (Article 31).
5. ☒ A copy of the International Application as filed (35 U.S.C. 371 (c) (2))
 - a. ☐ is attached hereto (required only if not communicated by the International Bureau).
 - b. ☒ has been communicated by the International Bureau.
 - c. ☐ is not required, as the application was filed in the United States Receiving Office (RO/US).
6. ☒ An English language translation of the International Application as filed (35 U.S.C. 371(c)(2)).
 - a. ☒ is attached hereto.
 - b. ☐ has been previously submitted under 35 U.S.C. 154(d)(4).
7. ☒ Amendments to the claims of the International Application under PCT Article 19 (35 U.S.C. 371 (c)(3))
 - a. ☐ are attached hereto (required only if not communicated by the International Bureau).
 - b. ☐ have been communicated by the International Bureau.
 - c. ☐ have not been made; however, the time limit for making such amendments has NOT expired.
 - d. ☒ have not been made and will not be made.
8. ☐ An English language translation of the amendments to the claims under PCT Article 19 (35 U.S.C. 371(c)(3)).
9. ☒ An oath or declaration of the inventor(s) (35 U.S.C. 371 (c)(4)).
10. ☒ An English language translation of the annexes to the International Preliminary Examination Report under PCT Article 36 (35 U.S.C. 371 (c)(5)).
11. ☐ A copy of the International Preliminary Examination Report (PCT/PEA/409).
12. ☒ A copy of the International Search Report (PCT/ISA/210).

Items 13 to 20 below concern document(s) or information included:

13. ☒ An Information Disclosure Statement under 37 CFR 1.97 and 1.98.
14. ☐ An assignment document for recording. A separate cover sheet in compliance with 37 CFR 3.28 and 3.31 is included.
15. ☒ A **FIRST** preliminary amendment.
16. ☐ A **SECOND** or **SUBSEQUENT** preliminary amendment.
17. ☐ A substitute specification.
18. ☐ A change of power of attorney and/or address letter.
19. ☐ A computer-readable form of the sequence listing in accordance with PCT Rule 13ter.2 and 35 U.S.C. 1.821 - 1.825.
20. ☐ A second copy of the published international application under 35 U.S.C. 154(d)(4).
21. ☐ A second copy of the English language translation of the international application under 35 U.S.C. 154(d)(4).
22. ☐ Certificate of Mailing by Express Mail
23. ☐ Other items or information:

Notice of Priority/PCT/IB/304

Form PTO-1449/Amended Sheets (Pages 8, 9 and 10)

Statement of Relevancy/Cited References (5)

U.S. APPLICATION NO. (IF KNOWN, SEE 37 CFR 1.53) <div style="font-size: 24pt; font-weight: bold;">10/089065</div>		INTERNATIONAL APPLICATION NO. <div style="font-weight: bold;">PCT/EP00/09630</div>		ATTORNEY'S DOCKET NUMBER <div style="font-weight: bold;">220355USOPCT</div>	
--	--	---	--	--	--

24. The following fees are submitted: BASIC NATIONAL FEE (37 CFR 1.492 (a) (1) - (5)) : <input type="checkbox"/> Neither international preliminary examination fee (37 CFR 1.482) nor international search fee (37 CFR 1.445(a)(2)) paid to USPTO and International Search Report not prepared by the EPO or JPO \$1040.00 <input checked="" type="checkbox"/> International preliminary examination fee (37 CFR 1.482) not paid to USPTO but International Search Report prepared by the EPO or JPO \$890.00 <input type="checkbox"/> International preliminary examination fee (37 CFR 1.482) not paid to USPTO but international search fee (37 CFR 1.445(a)(2)) paid to USPTO \$740.00 <input type="checkbox"/> International preliminary examination fee (37 CFR 1.482) paid to USPTO but all claims did not satisfy provisions of PCT Article 33(1)-(4) \$710.00 <input type="checkbox"/> International preliminary examination fee (37 CFR 1.482) paid to USPTO and all claims satisfied provisions of PCT Article 33(1)-(4) \$100.00 <div style="text-align: right; font-weight: bold;">ENTER APPROPRIATE BASIC FEE AMOUNT =</div>				CALCULATIONS PTO USE ONLY	
				<div style="display: flex; justify-content: space-between;"> \$890.00 </div>	
Surcharge of \$130.00 for furnishing the oath or declaration later than months from the earliest claimed priority date (37 CFR 1.492 (e)) <input type="checkbox"/> 20 <input type="checkbox"/> 30				<div style="display: flex; justify-content: space-between;"> \$0.00 </div>	
CLAIMS	NUMBER FILED	NUMBER EXTRA	RATE		
Total claims	13 - 20 =	0	x \$18.00	\$0.00	
Independent claims	3 - 3 =	0	x \$84.00	\$0.00	
Multiple Dependent Claims (check if applicable) <input type="checkbox"/>				\$0.00	
TOTAL OF ABOVE CALCULATIONS =				\$890.00	
<input type="checkbox"/> Applicant claims small entity status. See 37 CFR 1.27. The fees indicated above are reduced by 1/2.				\$0.00	
SUBTOTAL =				\$890.00	
Processing fee of \$130.00 for furnishing the English translation later than months from the earliest claimed priority date (37 CFR 1.492 (f)) <input type="checkbox"/> 20 <input type="checkbox"/> 30				\$0.00	
TOTAL NATIONAL FEE =				\$890.00	
Fee for recording the enclosed assignment (37 CFR 1.21 (h)). The assignment must be accompanied by an appropriate cover sheet (37 CFR 3.28, 3.31) (check if applicable) <input type="checkbox"/>				\$0.00	
TOTAL FEES ENCLOSED =				\$890.00	
				Amount to be:	\$
				refunded	
				charged	\$

a. ☒ A check in the amount of **\$890.00** to cover the above fees is enclosed.


b. ☐ Please charge my Deposit Account No. _____ in the amount of _____ to cover the above fees. A duplicate copy of this sheet is enclosed.

c. ☒ The Commissioner is hereby authorized to charge any additional fees which may be required, or credit any overpayment to Deposit Account No. **15-0030**. A duplicate copy of this sheet is enclosed.

d. ☐ Fees are to be charged to a credit card. **WARNING:** Information on this form may become public. Credit card information should not be included on this form. Provide credit card information and authorization on PTO-2038.

NOTE: Where an appropriate time limit under 37 CFR 1.494 or 1.495 has not been met, a petition to revive (37 CFR 1.137(a) or (b)) must be filed and granted to restore the application to pending status.

SEND ALL CORRESPONDENCE TO:



22850

SIGNATURE _____

William F. Beaumont
Norman F. Odion

REGISTRATION NUMBER 30,993

24,618

REGISTRATION NUMBER

April 5, 2002

DATE

220355US-0PCT

IN THE UNITED STATES PATENT & TRADEMARK OFFICE

IN RE APPLICATION OF :

KLAUS MUELLER-ENGEL : ATTN: APPLICATION DIVISION

SERIAL NO: NEW U.S. PCT APPLN :
(Based on PCT/EP00/09630)

FILED: HEREWITH :

FOR: CHEMICAL AND/OR PHYSICAL
TREATMENT OF MIXTURES
WHICH CONTAIN AT LEAST
ONE CHEMICAL COMPOUND
HAVING AT LEAST ONE
ETHYLENICALLY
UNSATURATED GROUPPRELIMINARY AMENDMENTASSISTANT COMMISSIONER FOR PATENTS
WASHINGTON, D.C. 20231

SIR:

Prior to examination on the merits, please amend the above-identified application as follows:

IN THE CLAIMS

Please amend the claims as shown on the marked-up copy following this amendment to read as follows:

3. (Amended) A process as claimed in claim 1, wherein the mixture to be treated is a mixture containing, as components, (meth)acrylic acid and an organic liquid having a higher boiling point than (meth)acrylic acid.

4. (Amended) A process as claimed in claim 1, wherein the mixture to be treated comprises $\geq 95\%$ by weight of (meth)acrylic acid.

5. (Amended) A process as claimed in claim 1, which is a rectification, extraction or absorption process.

6. (Amended) A process as claimed in claim 1, wherein R¹, R², R³ and R⁴ are either all methyl or all ethyl.

7. (Amended) A process as claimed in claim 1, wherein X is H.

8. (Amended) A process as claimed in claim 1, which is carried out at from 100 to 200°C.

9. (Amended) A process as claimed in claim 1, which is carried out at ≤ 100 mbar.

12. (Amended) A mixture as claimed in claim 10, wherein R¹, R², R³ and R⁴ are either all methyl or all ethyl.

REMARKS

Claims 1-13 are active in the present application. Claims 3-9 and 12 have been amended to remove multiple dependencies. No new matter is added. An action on the merits and allowance of claims is solicited.

Respectfully submitted,

OBLON, SPIVAK, McCLELLAND,
MAIER & NEUSTADT, P.C.



Norman F. Oblon
Attorney of Record
Registration No. 24,618

Stefan U. Koschmieder, Ph.D.
Registration No. 50,238



22850

(703) 413-3000
Fax #: (703) 413-2220
NFO/kst

I:\atty\SUKOS\220355us-pr.wpd

Marked-Up Copy
Serial No: _____
Amendment Filed on: _____
4-5-2002

IN THE CLAIMS

--3. (Amended) A process as claimed in claim 1 [or 2], wherein the mixture to be treated is a mixture containing, as components, (meth)acrylic acid and an organic liquid having a higher boiling point than (meth)acrylic acid.

4. (Amended) A process as claimed in [any of claims 1 to 3] claim 1, wherein the mixture to be treated comprises $\geq 95\%$ by weight of (meth)acrylic acid.

5. (Amended) A process as claimed in [any of claims 1 to 4] claim 1, which is a rectification, extraction or absorption process.

6. (Amended) A process as claimed in [any of claims 1 to 5] claim 1, wherein R^1 , R^2 , R^3 and R^4 are either all methyl or all ethyl.

7. (Amended) A process as claimed in [any of claims 1 to 6] claim 1, wherein X is H.

8. (Amended) A process as claimed in [any of claims 1 to 7] claim 1, which is carried out at from 100 to 200°C.

9. (Amended) A process as claimed in [any of claims 1 to 8] claim 1, which is carried out at ≤ 100 mbar.

12. (Amended) A mixture as claimed in claim 10 [or 11], wherein R^1 , R^2 , R^3 and R^4 are either all methyl or all ethyl.--

Chemical and/or physical treatment of mixtures which contain at least one chemical compound having at least one ethylenically unsaturated group

5

The present invention relates to a process for the chemical and/or physical treatment of mixtures which contain at least one chemical compound having at least one ethylenically unsaturated group.

10

Chemical compounds which contain at least one ethylenically unsaturated group (monomers) are generally known and are important starting compounds for the preparation of polymers (for example by free radical polymerization) which are used, inter alia, as adhesives or as binders.

15

In the preparation of monomers, e.g. (meth)acrylic acid (" (meth)acryl-" is used in this publication as an abbreviation for "acryl- or methacryl-"), esters of (meth)acrylic acid, nitriles of (meth)acrylic acid or styrene, it is constantly necessary to subject mixtures which contain at least one monomer to chemical and/or physical treatments in a manner known per se.

20

The esterification of (meth)acrylic acid with monohydric or polyhydric alkanols (cf. for example EP-A 463 434) or the treatment of (meth)acrylic acid-containing mixtures by rectification (cf. for example DE-A 19 810 962 or EP-A 648 732) may be mentioned by way of example.

25

The disadvantage of these known processes for the chemical and/or physical treatment of mixtures which contain at least one chemical compound having at least one ethylenically unsaturated group is that monomers tend to undergo undesired free radical polymerization, and said processes are therefore usually carried out in the presence of free radical polymerization inhibitors. Known free radical polymerization inhibitors of this type are, for example, nitroxyl radicals (compounds which have at least one >N-O- group) (cf. for example WO 9 921 893 and US-A 4 670 131).

30

35

However, even when free radical polymerization inhibitors are present, undesired free radical polymerization of monomers frequently cannot be ruled out, and it is for this reason that, in addition to polymerization inhibitors, substances which keep undesired polymer formed in suspension, i.e. which are intended to prevent formation of polymer deposits on, for example, container walls, column bottoms or evaporator surfaces, are also frequently added in processes for the chemical and/or physical

40

45

2

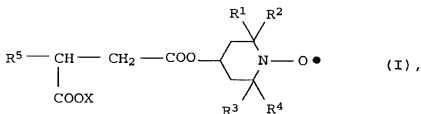
treatment of mixtures which contain at least one chemical compound having at least one ethylenically unsaturated group. Such substances are referred to as antifouling compositions (cf. for example US-A 3 271 296).

5

Against this background, it is an object of the present invention to provide processes for the chemical and/or physical treatment of mixtures which contain at least one chemical compound having at least one ethylenically unsaturated group, which processes are carried out in the presence of substances which form, on the one hand, excellent free radical polymerization inhibitors and, on the other hand, excellent antifouling compositions.

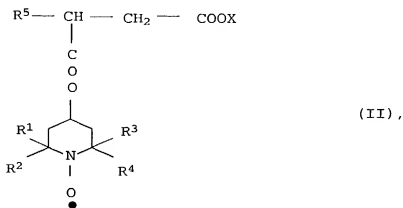
We have found that this object is achieved by a process for the chemical and/or physical treatment of mixtures which contain at least one chemical compound having at least one ethylenically unsaturated group, which is carried out in the presence of at least one compound of the formulae (I) and (II)

20



25

30



40

where X

R¹, R², R³ and R⁴,

is H, an alkali metal and/or ammonium independently of one another, are each C₁- to C₄-alkyl and is C₈- to C₃₀-alkyl.

45 R⁵

3

Compounds of the formulae (I) and (II) are disclosed, for example, in US-A 5 496 875 and are recommended there as intermediates for the preparation of light and heat stabilizers for polymers.

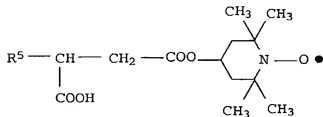
5

According to the invention, suitable alkali metals X are in particular Na and K. R¹, R², R³ and R⁴, independently of one another, may be methyl, ethyl, n-propyl, isopropyl, n-butyl, isobutyl or tert-butyl. Accordingly, compounds (I) and (II) in which all radicals R¹, R², R³ and R⁴ are methyl or in which all radicals R¹, R², R³ and R⁴ are ethyl are also suitable according to the invention. R⁵ may be, inter alia, C₁₅- to C₂₅-alkyl or C₁₇- to C₂₂-alkyl.

15 Preparation of compounds (I) and (II) is also described in US-A 5 496 875.

For the preparation of the compounds

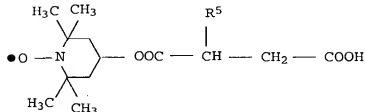
20



25

and

30



35

for example, the corresponding alkylsuccinic anhydrides can be reacted with 4-hydroxy-2,2,6,6-tetramethylpiperidine-N-oxyl (HTEMPO) at from 60 to 120°C. As a rule, the anhydride:N-oxyl molar ratio chosen for this purpose is from 0.8:1 to 1.5:1.

40 Preferably, the reaction is carried out in the absence of a solvent. Where solvents are used, suitable ones are, for example, aromatic and/or aliphatic hydrocarbons, such as toluene, xylene and cyclohexane, as well as diphenyl ether, dialkyl phthalates, dialkylacetamides and N-alkylpyrrolidones. The reaction time is as a rule 0.1-5 hours. The synthesis is advantageously carried out under anhydrous conditions.

The alkylsuccinic anhydrides used may be, for example, compounds such as tetrapropenylsuccinic anhydride (e.g. GP 103 from CONDEA), n-alkenylsuccinic anhydride having a number average molecular weight (M_n) of about 520 (e.g. GP 104 from CONDEA), or polyisobutenylsuccinic anhydride having an M_n of about 850 (e.g. GP 105 from CONDEA) or having an M_n of about 1400 (e.g. GP 106 from CONDEA).

In the novel process, possible compounds having at least one
10 ethylenically unsaturated group may be, for example, styrene,
butadiene, ethylene, vinyl ethers, vinyl esters, acrylic acid,
methacrylic acid, alkyl esters (in particular C₁- to C₈-alkyl) of
acrylic acid and methacrylic acid, methacrylonitrile,
acrylonitrile or N-vinylpyrrolidone.

15 The compounds I and II to be used concomitantly according to the invention are employed as a rule in amounts of from 50 to 1000 ppm by weight, based on the amount of the monomers contained in the mixture to be treated according to the invention. Of course, 20 the amount used may however also be up to 2000 or up to 3000 ppm by weight, on a corresponding basis. In suitable cases, however, it is of course also possible to use less than 50 ppm by weight.

The compounds (I) and (II) to be used according to the invention
25 are preferably chosen so that, when used in the required amount,
they are soluble in the mixture to be treated according to the
invention.

According to the invention, mixtures of compounds I and II are
30 generally used.

Of course, the compounds I and II can be used in the novel processes also as a mixture with other, known polymerization inhibitors and/or antifouling compositions. Suitable substances of this type include air, hydroquinone, hydroquinone monoethyl ether (MEHQ), paranitrosophenol, paramethoxyphenol, phenothiazine (PTZ), phenylenediamines, 4-hydroxy-2,2,6,6-tetramethylpiperidine-N-oxyl (HTEMPO), organic sulfonic acids (for example those published in EP-A 648 732), surfactants (for example those mentioned in DE-A 19810962) and all polymerization inhibitors stated in WO 9921893.

Advantageous combinations are, for example,

- 45 a) compounds I and II / PTZ;
b) compounds I and II / PTZ / MEHQ;
c) compounds I and II / PTZ / MEHQ / HTEMPO;

5

- d) compounds I and II / MEHQ / HTEMPO;
- e) compounds I and II / MEHQ;
- f) compounds I and II / MEHQ / HTEMPO.

- 5 The novel chemical treatment may be, for example, a conventional chemical reaction of a monomer with retention of the at least one ethylenically unsaturated group. The esterification of, for example, (meth)acrylic acid with alkanols has already been mentioned as an example. The novel physical treatment may be, for
- 10 example, an extraction, distillation, rectification, absorption or crystallization process.

- The mixtures to be treated according to the invention may be, for example, pure monomer mixtures as well as mixtures of monomers
- 15 and of substances other than monomers. As a rule, the weight fraction of the monomers in the mixtures to be treated according to the invention is at least 5 or at least 10 or at least 15 or 25 or 40% by weight.

- 20 In particular, the mixture to be treated according to the invention may comprise $\geq 95\%$ by weight of (meth)acrylic acid.

- The novel process is suitable, inter alia, for isolating, by rectification, (meth)acrylic acid from a mixture containing, as
- 25 main components, (meth)acrylic acid and an organic liquid having a higher boiling point than (meth)acrylic acid, as described in DE-A 19810962. The compounds I and II to be used according to the invention can be fed to the isolation by rectification at all points where WO 9921893 recommends the addition of a surfactant.
- 30 If required, they can be used together with surfactants. Frequently, the compounds I and II are added in solution in (meth)acrylic acid. Moreover, the novel procedure is also suitable in the case of the processes for the purification of crude (meth)acrylic acid by distillation, as described in EP-A
- 35 648 732. The compounds I and II to be used according to the invention can be employed alternatively to or together with the sulfonic acids and polymerization inhibitors used in EP-A 648 732.

40 Examples

- a) In each case 1 g of a crude acrylic acid which has been obtained by catalytic gas-phase oxidation of acroleine according to Example B1 of DE-A 4 302 991 and subsequent
- 45 working-up of the reaction gas mixture according to Example B1 of DE-A 2 136 396 and which has been stabilized with 300 ppm by weight of phenothiazine was mixed, under conditions of

6

saturation with air, with various amounts of different polymerization inhibitors and/or antifouling compositions in a test tube.

5 Thereafter, in each case 5 mg of azobisisobutyronitrile (free radical polymerization initiator) were added and the samples were heated open at 60°C in a water bath.

10 The time taken for the sample to begin to polymerize was then determined (detector: the liberated heat of polymerization). The times obtained as a function of added polymerization inhibitor and/or antifouling composition are shown in Table 1 below. The stated amounts are based on the total amount of the mixture. When no additional inhibitor or antifouling composition was added (i.e. the starting crude acrylic acid
15 was used alone), the time was 19 minutes.

Table 1

20	Composition added		Time (min)
	300 ppm by weight of GP 103 (from CONDEA)		19
	600 ppm by weight of GP 104 (from CONDEA)		20
	300 ppm by weight of HTEMPO		25
25	300 ppm by weight of GP 103 (from CONDEA) and 300 ppm by weight of HTEMPO		24
	600 ppm by weight of the reaction product of GP 103 (from CONDEA) with HTEMPO		24

It is noteworthy that the inhibitory effect of HTEMPO on the free radical polymerization is not impaired by the chemical bonding of
30 GP 103.

b) 137 g/h of a crude acrylic acid which had been obtained by catalytic gas-phase oxidation of acroleine according to Example B1 of DE-A 4 302 991 and subsequent working-up of the reaction gases according to Example B1 of DE-A-2 136 396 were
35 fed continuously, via the evaporator, into a glass rectification unit which was to be operated continuously and whose evaporator was a convection reboiler which was heated by means of a metallic, electrically heatable element,
40 1100 ppm by weight of aminoguanidine bicarbonate (as an aldehyde scavenger) and the polymerization inhibitors and/or antifouling compositions to be tested (cf. Table 2) having been added to said crude acrylic acid before it was fed into the evaporator. The temperature in the reboiler was 78°C and
45 the pressure at the top of the column was 100 mbar. The

7

column was 1.5 m long and was filled with raschig rings (5 mm, glass).

- 5 The mixture removed via the column and comprising low boilers contained in the crude acrylic acid, such as acetic acid and water, and small amounts of acrylic acid was condensed. 25 g/h of the condensate were removed and the remainder was recycled as reflux to the top of the column. To stabilize the column, a solution of 5000 ppm by weight of phenothiazine in pure acrylic acid was added at the top of said column
- 10 (20 ml/h). The bottom product substantially freed from the low boilers was removed from the evaporator under level control. During the rectification, deposition occurred on the heating element, the amount of which was weighed after an operating time of, in each case, 40 hours for each
- 15 polymerization inhibitor and/or antifouling composition to be tested. The results obtained are shown in Table 2.

Table 2

20

	Composition added	Deposition
	100 ppm by weight of HTEMPO	7.7 g
25	100 ppm by weight of HTEMPO and 100 ppm by weight of GP 103 (from CONDEA)	4.2 g
	200 ppm by weight of the reaction product of GP 103 (from CONDEA) with HTEMPO	1.4 g
	400 ppm by weight of the reaction product of GP 104 (from CONDEA) with HTEMPO	1.1 g
30	600 ppm by weight of the reaction product of GP 105 (from CONDEA) with HTEMPO	1.2 g

It is noteworthy that the compounds I and II to be used according to the invention are not only outstanding polymerization inhibitors but also excellent antifouling compositions.

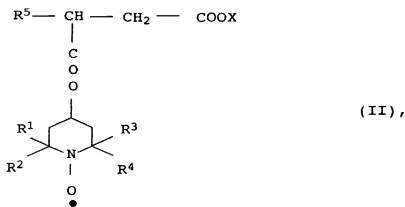
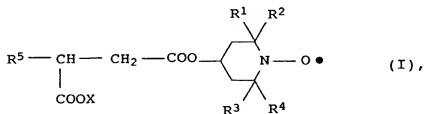
35

40

45

Amended claims:

1. A process for the chemical and/or physical treatment of a mixture which contains at least one chemical compound having at least one ethylenically unsaturated group, which is carried out in the presence of at least one compound of the formulae (I) and (II)

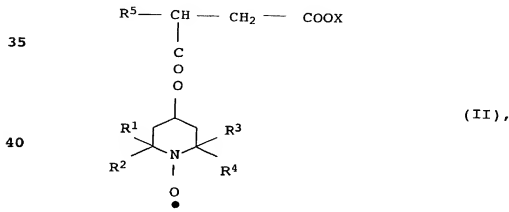
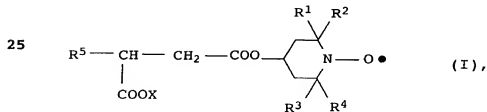


where X is H, an alkali metal and/or ammonium,
 R^1 , R^2 , R^3 and R^4 , independently of one another, are each C_1 - to C_4 -alkyl and
 R^5 is C_8 - to C_{30} -alkyl.

2. A process as claimed in claim 1, wherein the at least one chemical compound having at least one ethylenically unsaturated group is acrylic acid, methacrylic acid, acrylonitrile, methacrylonitrile, styrene, an ester of acrylic acid and/or an ester of methacrylic acid.
3. A process as claimed in claim 1 or 2, wherein the mixture to be treated is a mixture containing, as components, (meth)acrylic acid and an organic liquid having a higher boiling point than (meth)acrylic acid.

9

4. A process as claimed in any of claims 1 to 3, wherein the mixture to be treated comprises $\geq 95\%$ by weight of (meth)acrylic acid.
5. A process as claimed in any of claims 1 to 4, which is a rectification, extraction or absorption process.
6. A process as claimed in any of claims 1 to 5, wherein R^1 , R^2 , R^3 and R^4 are either all methyl or all ethyl.
7. A process as claimed in any of claims 1 to 6, wherein X is H.
8. A process as claimed in any of claims 1 to 7, which is carried out at from 100 to 200°C.
9. A process as claimed in any of claims 1 to 8, which is carried out at ≤ 100 mbar.
10. A mixture which contains at least one chemical compound having at least one ethylenically unsaturated group and at least one compound of the formulae (I) and (II)



10

where X is H, an alkali metal and/or ammonium,
R¹, R², R³ and R⁴, independently of one another, are each
C₁- to C₄-alkyl and
R⁵ is C₈- to C₃₀-alkyl.

- 5
11. A mixture as claimed in claim 10, in which the at least one chemical compound having at least one ethylenically unsaturated group is selected from the group consisting of acrylic acid, methacrylic acid, acrylonitrile,
10 methacrylonitrile, styrene, esters of acrylic acid and esters of methacrylic acid.
12. A mixture as claimed in claim 10 or 11, wherein R¹, R², R³ and R⁴ are either all methyl or all ethyl.
- 15
13. A process for the chemical and/or physical treatment of a mixture which contains at least one chemical compound having at least one ethylenically unsaturated group, which is carried out in the presence of at least one compound which is
20 obtainable by reacting an alkylsuccinic anhydride having a number average molar mass (M_n) of from 212 to about 1400 with 4-hydroxy-2,2,6,6-tetramethylpiperidin-N-onyl.

25

30

35

40

45

11

Chemical and/or physical treatment of mixtures which contain at least one chemical compound having at least one ethylenically unsaturated group

5

Abstract

Mixtures containing monomers having at least one ethylenically unsaturated group are chemically and/or physically treated in the presence of the reaction products of alkylsuccinic anhydrides and stable N-oxyl radicals having a hydroxyl group.

15

20

25

30

35

40

45

(12) NACH DEM VERTRAG ÜBER DIE INTERNATIONALE ZUSAMMENARBEIT AUF DEM GEBIET DES
PATENTWESENS (PCT) VERÖFFENTLICHTE INTERNATIONALE ANMELDUNG

(19) Weltorganisation für geistiges Eigentum
Internationales Büro



(43) Internationales Veröffentlichungsdatum
12. April 2001 (12.04.2001)

PCT

(10) Internationale Veröffentlichungsnummer
WO 01/25173 A1

(51) Internationale Patentklassifikation: C07B 63/04,
C09K 15/30, C07C 51/50

Mundenheimer Strasse 170, 67061 Ludwigshafen (DE).
SCHRÖDER, Jürgen [DE/DE]; Niedererdstrasse 20,
67071 Ludwigshafen (DE).

(21) Internationales Aktenzeichen: PCT/EP00/09630

(74) Gemeinsamer Vertreter: BASF AKTIENGE-
SELLSCHAFT; 67056 Ludwigshafen (DE).

(22) Internationales Anmeldedatum:
2. Oktober 2000 (02.10.2000)

(81) Bestimmungsstaaten (national): BR, US.

(25) Einreichungssprache: Deutsch

(84) Bestimmungsstaaten (regional): europäisches Patent (AT,
BE, CH, CY, DE, DK, ES, FI, FR, GB, GR, IE, IT, LU, MC,
NL, PT, SE).

(26) Veröffentlichungssprache: Deutsch

(30) Angaben zur Priorität:
199 47 868.6 5. Oktober 1999 (05.10.1999) DE

Veröffentlicht:

— Mit internationalem Recherchenbericht.
— Vor Ablauf der für Änderungen der Ansprüche geltenden
Frist; Veröffentlichung wird wiederholt, falls Änderungen
eintreffen.

(71) Anmelder (für alle Bestimmungsstaaten mit Ausnahme
von US): BASF AKTIENGESSELLSCHAFT [DE/DE];
67056 Ludwigshafen (DE).

Zur Erklärung der Zweibuchstaben-Codes, und der anderen
Abkürzungen wird auf die Erklärungen ("Guidance Notes on
Codes and Abbreviations") am Anfang jeder regulären Ausgabe
der PCT-Gazette verwiesen.

(72) Erfinder; und

(75) Erfinder/Anmelder (nur für US): MÜLLER-EN-
GEL, Klaus, Joachim [DE/DE]; Bahnhofstrasse 82,
76297 Sautensee (DE). NESTLER, Gerhard [AT/DE];

(54) Title: TREATMENT OF MIXTURES THAT CONTAIN AT LEAST ONE COMPOUND WITH AT LEAST ONE ETHYLENICALLY UNSATURATED GROUP

(54) Bezeichnung: BEHANDLUNG VON GEMISCHEN, DIE WENIGSTENS EINE ETHYLENISCH UNGESÄTTIGTE VER-
BINDUNG ENTHALTEN

(57) Abstract: The invention relates to the chemical and/or physical treatment of mixtures that contain monomers having at least one ethylenically unsaturated group in the presence of the reaction products of alkyl succinic acid anhydrides and stable N-oxyl radicals that have one hydroxy group.

(57) Zusammenfassung: Die chemische und/oder physikalische Behandlung von wenigstens eine ethylenisch ungesättigte Gruppe aufweisenden Monomere enthaltenden Gemischen im Beisein der Umsetzungsprodukte von Alkylbernsteinsäureanhydriden und ein Hydroxygruppe aufweisenden stabilen N-Oxyl-Radikalen.

WO 01/25173 A1

Declaration and Power of Attorney for Patent Application

Erklärung für Patentanmeldungen mit Vollmacht

German Language Declaration

Als nachstehend benannter Erfinder erkläre ich hiermit an Eides Statt:

As a below named inventor, I hereby declare that:

Dass mein Wohnsitz, meine Postanschrift und meine Staatsangehörigkeit den im nachstehenden nach meinem Namen aufgeführten Angaben entsprechen, dass ich nach bestem Wissen der ursprüngliche, erste und alleinige Erfinder (falls nachstehend nur ein Name angegeben ist) oder ein ursprünglicher, erster und Miterfinder (falls nachstehend mehrere Namen aufgeführt sind) des Gegenstandes bin, für den dieser Antrag gestellt wird und für den ein Patent für die Erfindung mit folgendem Titel beantragt wird:

My residence, post office address and citizenship are as stated next to my name.

I believe I am the original, first and sole inventor (if only one name is listed below) or an original, first and joint inventor (if plural names are listed below) of the subject matter which is claimed and for which a patent is sought on the invention entitled:

CHEMICAL AND/OR PHYSICAL TREATMENT OF MIXTURES WHICH
CONTAIN AT LEAST ONE CHEMICAL COMPOUND HAVING AT LEAST
ONE ETHYLENICALLY UNSATURATED GROUP

deren Beschreibung:

the specification of which:

☐ ist beigelegt

☐ is attached hereto

☐ wurde angemeldet am _____

☒ was filed on *02 October 2000*

unter der US-Anmeldenummer oder unter der Internationalen Anmeldenummer im Rahmen des Vertrages über die Zusammenarbeit auf dem Gebiet des Patentwesens (PCT)

as United States Application Number or PCT International Application Number

_____ und am _____

PCT/EP/00/09630 _____
and was amended on _____

_____ abgeändert (falls zutreffend).

_____ (if applicable).

Ich bestätige hiermit, dass ich den Inhalt der oben angegebenen Patentanmeldung, einschließlich der Ansprüche, die eventuell durch einen oben erwähnten Zusatzantrag abgeändert wurde, durchgesehen und verstanden habe.

I hereby state that I have reviewed and understand the contents of the above identified specification, including the claims, as amended by any amendment referred to above.

Ich erkenne meine Pflicht zur Offenbarung jeglicher Informationen an, die zur Prüfung der Patentfähigkeit in Einklang mit Titel 37, Code of Federal Regulations, § 1.56 von Belang sind.

I acknowledge the duty to disclose information which is material to patentability as defined in Title 37, Code of Federal Regulations, § 1.56.

German Language Declaration

Ich beanspruche hiermit ausländische Prioritätsvorteile gemäß Titel 35, US-Code, § 119(a)-(d), bzw. § 365(b) aller unten aufgeführten Auslandsanmeldungen für Patente oder Erfinderrkunden, oder § 365(a) aller PCT internationalen Anmeldungen, welche wenigstens ein Land außer den Vereinigten Staaten von Amerika benennen, und habe nachstehend durch Ankreuzen sämtlicher Auslandsanmeldungen für Patente bzw. Erfinderrkunden oder PCT internationale Anmeldungen angegeben, deren Anmeldetag dem der Anmeldung, für welche Priorität beansprucht wird, vorangeht.

Prior foreign application(s)
(Frühere ausländische Anmeldungen)

19947868.6	Germany
(Number)	(Country)
(Nummer)	(Land)

Ich beanspruche hiermit Prioritätsvorteile unter Titel 35, US-Code, § 119(e) aller US-Hilfsanmeldungen wie unten aufgezählt.

(Application No.)	(Filing Date)
(Aktenzeichen)	(Anmeldetag)

Ich beanspruche hiermit die mir unter Titel 35, US-Code, § 120 zustehenden Vorteile aller unten aufgeführten US-Patentanmeldungen bzw. § 365(c) aller PCT internationalen Anmeldungen, welche die Vereinigten Staaten von Amerika benennen, und erkenne, insofern der Gegenstand eines jeden früheren Anspruchs dieser Patentanmeldung nicht in einer US-Patentanmeldung, bzw. PCT internationalen Anmeldung in einer gemäß dem ersten Absatz von Titel 35, US-Code, § 112 vorgeschriebenen Art und Weise offenbart wurde, meine Pflicht zur Offenbarung jeglicher Informationen an, die zur Prüfung der Patentfähigkeit in Einklang mit Titel 37, Code of Federal Regulations, §1.56 von Belang sind und die im Zeitraum zwischen dem Anmeldetag der früheren Patentanmeldung und dem nationalen oder im Rahmen des Vertrages über die Zusammenarbeit auf dem Gebiet des Patentwesens (PCT) gültigen internationalen Anmeldetages bekannt geworden sind.

(Application No.)	(Filing Date)
(Aktenzeichen)	(Anmeldetag)

(Application No.)	(Filing Date)
(Aktenzeichen)	(Anmeldetag)

I hereby claim foreign priority under Title 35, United States Code, § 119(a)-(d) or §365(b) of any foreign application(s) for patent or inventor's certificate, or § 365(a) of any PCT International application which designated at least one country other than the United States, listed below, and have also identified below, by checking the box, any foreign application for patent or inventor's certificate, or PCT International application having a filing date before that of the application on which priority is claimed.

Priority claimed

Priorität
beansprucht

05 October 1999	[x]	[]
(Day/Month/Year filed)	Yes	No
(Tag/Monat/Jahr der Anmeldung)	Ja	Nein

I hereby claim the benefit under Title 35, United States Code, § 119(e) of any United States provisional application(s) listed below.

(Application No.)	(Filing Date)
(Aktenzeichen)	(Anmeldetag)

I hereby claim the benefit under Title 35, United States Code, § 120 of any United States application(s), or § 365(c) of any PCT International application designating the United States, listed below and, insofar as the subject matter of each of the claims of this application is not disclosed in the prior United States or PCT International application in the manner provided by the first paragraph of Title 35, United States Code, § 112, I acknowledge the duty to disclose information which is material to patentability as defined in Title 37, Code of Federal Regulations, §1.56 which became available between the filing date of the prior application and the national or PCT International filing date of this application.

(Status) (patented, pending, abandoned)
(Status) (patentiert, schwebend, aufgegeben)

(Status) (patented, pending, abandoned)
(Status) (patentiert, schwebend, aufgegeben)

German Language Declaration

Ich erkläre hiermit, dass alle in der vorliegenden Erklärung von mir gemachten Angaben nach bestem Wissen und Gewissen der Wahrheit entsprechen, und ferner, dass ich diese eidesstattliche Erklärung in Kenntnis dessen ablege, dass wissentlich und vorsätzlich falsche Angaben oder dergleichen gemäß § 1001, Titel 18 des US-Code strafbar sind und mit Geldstrafe und/oder Gefängnis bestraft werden können und dass derartige wissentlich und vorsätzlich falsche Angaben die Rechtswirksamkeit der vorliegenden Patentanmeldung oder eines aufgrund deren erteilten Patentes gefährden können.

VERTRETUNGSVOLLMACHT: Als benannter Erfinder beauftrage ich hiermit den (die) nachstehend aufgeführten Patentanwalt (Patentanwälte) und/oder Vertreter mit der Verfolgung der vorliegenden Patentanmeldung sowie mit der Abwicklung aller damit verbundenen Angelegenheiten vor dem US-Patent- und Markenamt: *(Name(n) und Registrationsnummer(n) auflisten)*

I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

POWER OF ATTORNEY: As a named inventor, I hereby appoint the following attorney(s) and/or agent(s) to prosecute this application and transact all business in the Patent and Trademark Office connected therewith: *(list name and registration number)*



022850

Postanschrift:

Send Correspondence to:



022850

Telefonische Auskünfte:
(Name und Telefonnummer)

Direct Telephone calls to: *(name and telephone number)*

(703) 413-3000

DECLARATION

Page 4 of 4

0050/050783

100
Klaus Joachim Müller-Engel

NAME OF INVENTOR

Klaus Joachim Müller-Engel

Signature of Inventor

Date

1/18

October 19, 2000

Bahnhofstr.82

76297 Stutensee DEU

Germany

Citizen of: Germany

Post Office Address: same as residence

200
Gerhard Nestler

NAME OF INVENTOR

Gerhard Nestler

Signature of Inventor

Date

October 19, 2000

Mundenheimer Str.170

67061 Ludwigshafen DEU

Germany

Citizen of: Austria

Post Office Address: same as residence

300
Jürgen Schröder

NAME OF INVENTOR

Jürgen Schröder

Signature of Inventor

Date

October 19, 2000

Niedererdstr.20

67071 Ludwigshafen DEU

Germany

Citizen of: Germany

Post Office Address: same as residence